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Dynamic modelling of nitrous oxide emissions from three Swedish full-scale sludge liquor treatment systems

E. Lindblom[1,2], M. Arnell[1,3], X. Flores-Alsina[4], F. Stenström[5,6], D.J.I. Gustavsson[7,8], U. Jeppsson[1]

[1] Div. of Industrial Electrical Engineering and Automation (IEA), Dept of Biomedical Engineering, Lund University, P.O. Box 118, SE-221 00 Lund, Sweden (erik.lindblom@iea.lth.se)
[2] Stockholm Vatten AB, SE-106 36 Stockholm, Sweden (erik.lindblom@stockholmvatten.se)
[3] Urban Water Management Sweden AB, Gjuterigatan 1D, SE-582 73 Linköping, Sweden
[4] Dept of Chemical and Biochemical Engineering, Technical University of Denmark, Building 229, DK-2800 Lyngby, Denmark
[5] Water and Environmental Engineering, Dept of Chemical Engineering, Lund University, P.O. Box 118, SE-221 00 Lund, Sweden
[6] VA-Ingenjörerna AB, Trädgårdsgatan 12, SE-702 12 Örebro, Sweden
[7] VA SYD, P.O. Box 191, SE-201 21 Malmö, Sweden
[8] Sweden Water Research, Ideon Science Park, Scheelevägen 15, SE-223 70 Lund, Sweden

Abstract: The objective of this paper is to model the dynamics and validate the results of nitrous oxide (N_2O) emissions from three Swedish full-scale nitrifying/denitrifying, nitritation and anammox systems treating anaerobic digester sludge liquor. The Activated Sludge Model No. 1 is extended in order to describe N_2O production by both heterotrophic and autotrophic bacteria. In addition, mass transfer equations are implemented to characterize the dynamics of N_2O in the water and the gas phases. The biochemical model is simulated for two hydraulic patterns: 1) a sequencing batch reactor (SBR); and, 2) a moving-bed biofilm reactor (MBBR). Preliminary results show that the calibrated model is partly capable of reproducing the behaviour of N_2O as well as the nitritation/nitrification/denitrification dynamics. However, the results emphasize that more work is required before N_2O emissions from sludge liquor treatment plants can be generally predicted with certainty by simulations. Continued efforts should focus on determining the switching conditions for different N_2O formation pathways and, if full-scale data is used, modelling of the measurement devices might improve the conclusions that can be drawn.

Keywords: ASMN; Autotrophic denitrification; Greenhouse gases; Heterotrophic denitrification; Modelling; Sludge liquor treatment

Introduction

Efficient municipal wastewater treatment plant (WWTP) engineering and operation call for plant-wide process understanding, which can be summarized as mathematical models. Recent research have shown that some "optimal" WWTP operational strategies, e.g. operation with intermittent aeration and/or low dissolved oxygen (DO) set-points, might be "sub-optimal" in certain respects because of the risk for elevated emission of the undesired greenhouse gas nitrous oxide (N_2O). This is possibly due to lack of knowledge and the inability of WWTP simulators to realistically describe this effect. Determining the best actions for mitigation of N_2O emissions from full-scale WWTP processes is therefore a major problem.

Based on new knowledge of the biological mechanisms of N_2O production (e.g. Kampschreur *et al.*, 2009), recent efforts have been made to capture the production and emissions of N_2O and integrate these processes with the traditional ASM models (Hiatt and Grady, 2008; Mampaey *et al.*, 2013; Ni *et al.*, 2013; Pan *et al.*, 2014). The aim is to increase the understanding of the N_2O production mechanisms and eventually to allow for mitigation and control strategies taking the potential trade-offs between effluent water quality, resource consumption and greenhouse gas production in full-scale plants into consideration and optimize and evaluate such actions by means of simulations.

In this study, a biological process model that includes N_2O production in processes treating sludge liquors from anaerobic digestion of municipal primary and secondary sludge has been developed,

implemented and evaluated to test if a combination of the above-mentioned models can be calibrated and validated using the full-scale data.

Material and Methods

Full-scale data sets

The model is calibrated to reproduce the data sets from three Swedish full-scale systems denoted *SBR_N/DN*, *SBR_NO2* and *MBBR_AMX*:

- ***SBR_N/DN:*** N_2O measurements performed by Stenström *et al.* (2014), who investigated a nitrification(N)-denitrification(DN) SBR process at Slottshagen WWTP (Norrköping, Sweden);
- ***SBR_NO2:*** N_2O measurements performed by Gustavsson and la Cour Jansen (2011), who investigated a *nitritation only* SBR process at Sjölunda WWTP (Malmö, Sweden); and,
- ***MBBR_AMX:*** N_2O measurements performed by Yang *et al.* (2013), who investigated a one stage nitritation-anammox moving-bed biofilm reactor process at Hammarby-Sjöstad pilot plant (Stockholm, Sweden).

The three case studies involve anaerobic digestion sludge liquor treatment and all include measurements of traditional wastewater variables (online and grab samples) and online measurements of N_2O (water and/or gas phase). All measurement results have not been considered in the present study, the reader is referred to the original papers for further details of the experiments.

Mathematical models

Considering the experimental data of the three case studies, a biological process model including heterotrophic ($X_{B,H}$) and ammonia oxidizing bacteria (X_{AOB}) denitrification was hypothesized to be able to describe the measurements. The model was initially based on the ideas summarized in Hiatt and Grady (2008). This model (ASMN) extends the well-recognized ASM1 (Henze *et al.*, 2000) with two nitrifying populations: X_{AOB} and nitrite oxidizing bacteria (X_{NOB}), using ammonia (S_{NH3}) and nitrous acid (S_{HNO2}), respectively, as substrates. Sequential 4-step heterotrophic denitrification of nitrate (S_{NO3}) to nitrogen gas (N_2) via nitrite (S_{NO2}), nitric oxide (S_{NO}) and S_{N2O} is also included. However, the ASMN model does not include AOB denitrification, which, as pointed out by Gustavsson and la Cour Jansen (2011) and Stenström *et al.* (2014) amongst others, potentially is a governing process for N_2O formation in biological WWTPs. Therefore the N_2O production mechanisms recently proposed by Mampaey *et al.* (2013) were included in the model. Thereby, X_{AOB} are also capable of reducing S_{HNO2} to S_{NO} and further into S_{N2O}. The assumed reaction rates for X_{AOB} denitrification [g $N \cdot m^{-3} \cdot d^{-1}$)] are shown in Eq. 1 and Eq. 2:

$$r_{NO,AOB,den} = f_{DNT,A} \cdot \frac{\mu_{AOB}}{Y_{AOB}} \cdot \left(\frac{S_{O,AOB}}{K_{O,AOB} + S_{O,AOB}} \right) \cdot \left(\frac{S_{NH3,AOB}}{K_{NH3,AOB} + S_{NH3}} \right) \cdot \left(\frac{S_{HNO2}}{K_{HNO2,AOB} + S_{NO}} \right) \quad (1)$$

$$r_{N2O,AOB,den} = \frac{\mu_{AOB}}{Y_{AOB}} \cdot \left(\frac{S_{O,AOB}}{K_{O,AOB} + S_{O,AOB}} \right) \cdot \left(\frac{S_{NH3,AOB}}{K_{NH3,AOB} + S_{NH3}} \right) \cdot \left(\frac{S_{NO}}{K_{NO,AOB} + S_{NO}} \right) \quad (2)$$

In the original model, the same half-saturation coefficients $K_{O,AOB}$ and $K_{NH3,AOB}$ were assumed for X_{AOB} aerobic ammonia oxidation and X_{AOB} denitrification. The parameters $K_{HNO2,AOB}$ and $K_{NO,AOB}$ are unique for X_{AOB} denitrification. Finally, growth and decay processes of anammox active biomass (X_{AMX}) following Hao *et al.* (2002) were included in the biological process model. X_{AMX} convert S_{NH4} and S_{NO2} to mainly nitrogen gas and also S_{NO3} in the absence of oxygen.

Stripping (mass transfer) equations for the gases were implemented as in Foley *et al*. (2011). In the three case studies, the monitored dissolved oxygen (DO) concentration is used as the input to a

controller that adjusts the k_La_{O2} of the modelled system. The applied diffusivities of N_2O and O_2 are $1.77 \cdot 10^{-9}$ m^2/s and $2.12 \cdot 10^{-9}$ m^2/s, respectively, yielding $k_La_{N2O}=0.91 \cdot k_La_{O2}$. The simulated flux of N_2O in the offgas (F_{N2O} [kg $N \cdot d^{-1}$]), which is used to validate the model behaviour with the measured emissions, is then given by $F_{N2O}=k_La_{N2O} \cdot S_{N2O} \cdot V_{AER}$, with V_{AER} [m^3] denoting the aerated water volume. The simulated k_La_{O2} values are within the range of 300 to 600 d^{-1}. Thus, the half-life of possibly accumulated S_{N2O} during stripping is only a few minutes. Any long-term dynamic changes should therefore be due to variations of the biological reaction rates. However, the stripping/flux equation might represent an overly simplified version of reality. For example, the retention time of the bubbles in the reactor, the measurement devices and stripping during non-aerated conditions have not been taken into account.

The reactive settler model developed within the BSM framework (Flores-Alsina *et al.*, 2012) was expanded with variable layer heights (e.g. during filling) and layer mixing (e.g. during aeration) to describe the SBR behaviour of *SBR_N/DN* and *SBR_NO2* systems.

The biofilm model, used to model the MBBR anammox system (*MBBR_AMX*), was inspired by the implementation in the commercial software platform WEST 3.7.3 (DHI, 2011). According to this model, the bulk water volume is separated from the biofilm, which in turn is divided into 10 layers. Soluble components are transported by diffusion between the biofilm layers and bulk, proportionally to the concentration gradients. Particulate material attaches to the outermost layer of the biofilm and detachment occurs from all layers as the biofilm thickness exceeds a user defined maximum value.

Results and Discussion

Nitrification/denitrification sequencing batch reactor, case SBR_N/DN

Recorded DO and pH values (**Fig. 1A & D**) as well as flow rate data were directly used as model inputs. The process temperature was constant (30.3 ºC).

During the measurement period of 16 hours used for model calibration the NH_4^+-N load to the SBR plant was 180 kg $N \cdot d^{-1}$. The *SBR_N/DN* cycle of 8 hours starts with 3.5 hours of anoxic denitrification including 2 hours of filling. Initially, the accumulation rate of S_{N2O} is almost equal to the denitrification rate of S_{NO3} indicating that the final step of heterotrophic denitrification is inhibited (**Fig. 1G**). At t=1.5 h ethanol is dosed to the process and S_{N2O} is immediately reduced. To model these observations with ASMN the heterotrophic N_2O denitrification process without ethanol must be almost completely inhibited. The original ASMN inhibition term for S_{NO} was replaced by S_{NO2} inhibition (Zhou *et al.*, 2008) since no information of S_{NO} concentrations was available. Despite several attempts this drastic shift between complete and no inhibition because of a low availability of readily biodegradable substrate (S_S), could not be captured by the original ASMN model and motivated an extension by an additional model component representing ethanol, $S_{S,EtOH}$ [g $COD \cdot m^{-3}$]. This state variable was assumed to affect the process in the same way as S_S, with the exception that the half-saturation coefficient ($K_{SEtOH,5}$) for heterotrophic growth with N_2O as electron acceptor is set to a low value (1 g $COD \cdot m^{-3}$). For the same process, but with S_S from the influent sludge liquor as substrate, the half-saturation coefficient (K_{S5}) was given a high value (100 g $COD \cdot m^{-3}$) to model the inhibition without external carbon. According to the simulation results, S_{N2O} starts to accumulate again as ethanol is consumed, a phenomenon that was not measured and indicates that separated growth on internal and added substrates is not necessarily the actual process governing the S_{N2O} formation. Other concepts for modelling the denitrification process has recently been presented by Pan *et al*. (2014) and further experimental work and model calibration are required to fully describe the observed behaviour.

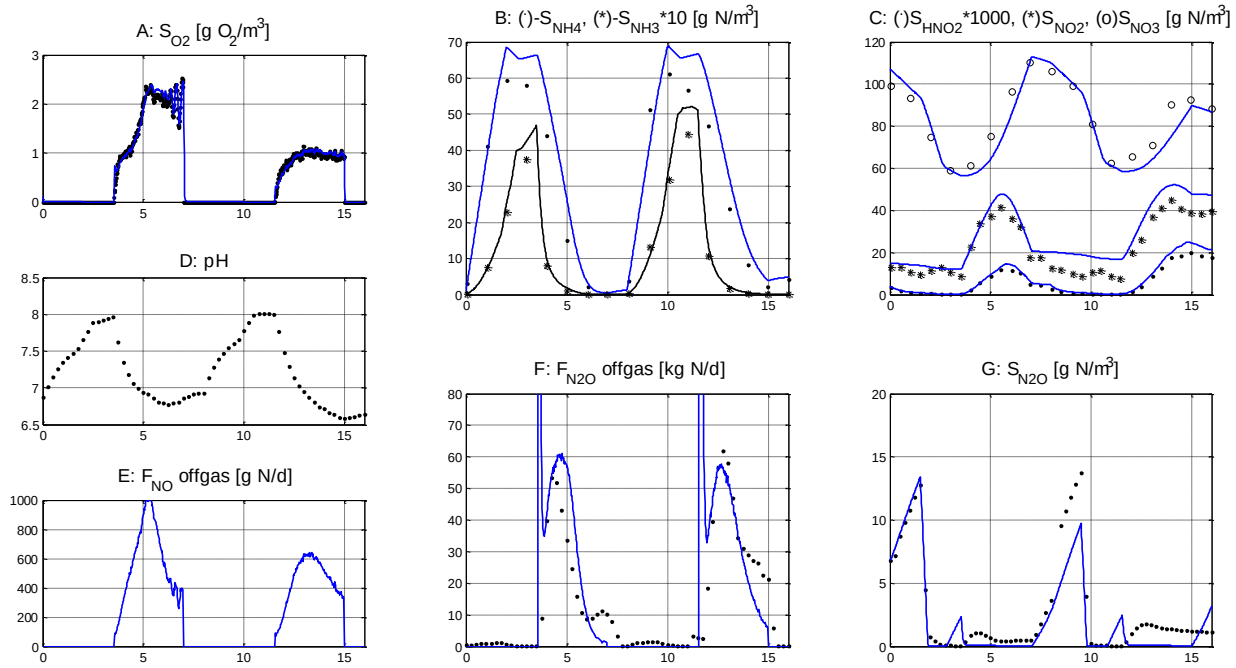

Figure 1. Measured (markers) and simulated (solid lines) concentrations and mass flows for the nitrification/ denitrification SBR process (*SBR_N/DN)*. The x-axes show time in hours.

Oxidation of S_{NH3} starts instantly when aeration is initiated at t=3.5 h (**Fig. 1A & B**). The associated N_2O emissions are shown in **Fig. 1F**. A sharp peak in the simulated emission is seen at the start of the aeration due to stripping of the in part faulty prediction of anoxic N_2O accumulation. The maximum N_2O emission is reached after 1 hour of aeration although absence of accumulated S_{N2O} from the preceding anoxic phase. Thus the emission is mainly due to N_2O production during aerobic conditions. The X_{AOB} affinity coefficient for S_{HNO2} appears relatively low since S_{HNO2} peaks after 2.5 hours of aeration (**Fig. 1C**). As will be shown in the next case study as well, the studied emission seems to be correlated to S_{NH3} and in the model this has been accounted for by choosing a separate half-saturation coefficient for S_{NH3} during AOB denitrification, $K_{NH3,AOB,DN}$ [g $N \cdot m^{-3}$]. The used value is 7 times higher compared to the value of $K_{NH3,AOB}$ (0.053 g $N \cdot m^{-3}$) for aerobic ammonia oxidation, see **Table 1**.

Nitritation only sequencing batch reactor, case SBR_NO2

During the measurement period of 24 hours used for model calibration the NH_4^+-N load to the SBR plant was 710 kg $N \cdot d^{-1}$. The temperature in the SBR process was similar to *SBR_N/DN*, 31.7 ºC. An important difference is, however, that in *SBR_NO2* the pH was controlled at 6.8. The *SBR_NO2* cycle of 6 hours starts with aeration and filling. S_{NH4} increases until filling stops after 1.5 hours (**Fig. 2B**). During the subsequent aerobic batch mode phases, the nitritation process proceeds until aeration is switched off. A fixed constant airflow is applied during each cycle but the total length of the aerated phases is varied. This also means that the time periods for the anoxic settling phases that make up the end of each cycle vary.

The SBR process had been operated for nitritation only during several months prior to this case study and the activated sludge was therefore enriched with X_{AOB}. From the DO data (**Fig. 2A**) it is seen that the oxygen demand of the sludge decreases when S_{NH4} decreases below 50 g $N \cdot m^{-3}$, which was modelled by a high half-saturation coefficient ($K_{NH3,AOB}$) value of 1.1 g $N \cdot m^{-3}$.

The N_2O emissions reach 30-75 kg $N \cdot d^{-1}$ and decrease to 10-15 kg $N \cdot d^{-1}$ at the end of the aerobic phases (**Fig. 2E**). In the most extreme cycle (#4, t=18-24 h), the emission at the end of the aerated phase is only 20% of the maximum emission during that same phase. The sharp simulated peaks at the beginning of each phase (not reflected in the measurement data) are due to stripping of accumulated S_{N2O}. Consequently, as stripping according to the model occurs fast, the decrease in

N_2O production throughout the aeration phase must be explained by aerobic biological N_2O production, which is entirely determined by X_{AOB} denitrification of S_{NO}.

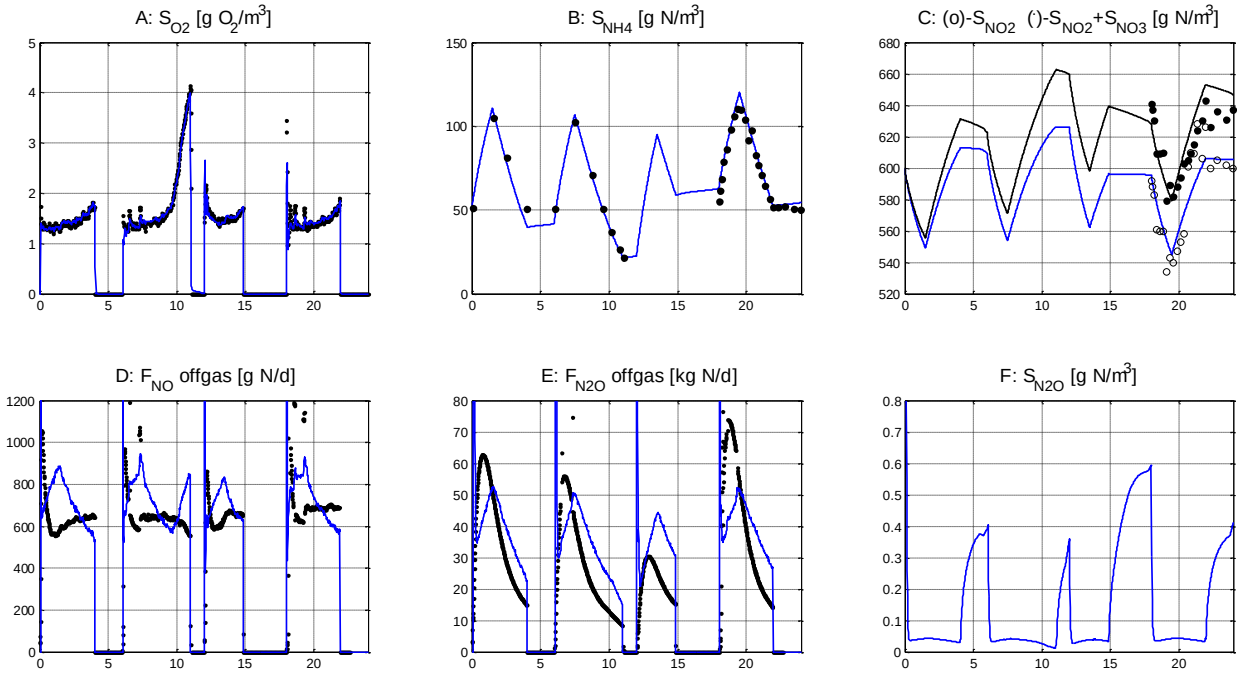

Figure 2. Measured (markers) and simulated (solid lines) concentrations and mass flows for the nitritation only SBR process (*SBR_NO2)*. The x-axes show time in hours.

Opposite to the *SBR_N/DN* case this study includes measurements of NO offgas concentrations, which were relatively stable. This is reflected by the almost constant calculated NO emissions shown in **Fig. 2D**. Considering Eq. 2, S_{NO} does therefore not explain the dynamic N_2O emissions. S_{NO2} in the process varies around 500-600 g $N \cdot m^{-3}$ during the nitritation phase (**Fig. 2C**) and since the concentrations are high they are believed not to represent any major cause for the varying N_2O emissions. S_O actually increases throughout the aeration phase, a phenomenon that according to the model could increase the N_2O production (in contrast to the observations).

The attempt to fit the Mampaey model (Mampaey *et al.*, 2013) to the measurement data is not successful and requires the inclusion of – as was also done for *SBR_N/DN* – a unique S_{NH3} half-saturation coefficient for AOB denitrification ($K_{NH3,AOB,DN}$). By choosing a high value (7 g $N \cdot m^{-3}$) the S_{NH3} dependency changes towards a linear relation and part of the dynamics can be modelled.

In the original paper describing the experimental data (Gustavsson and Jansen, 2011), a linear relation between the length of the anoxic phase and emitted mass of N_2O was proposed. The implemented model can be adjusted to explain this phenomenon as seen in the varying peak S_{N2O} concentrations before aeration (**Fig. 2F**). However, as already noted, the sharp peaks in the simulated emissions due to stripping were not experimentally supported.

The overall conclusion based on the reasoning above, and several attempts of simulating the model with various parameter sets, is that the ASMN/Mampaey model may not be feasible for explaining the complete dynamics of nitrous oxide emissions from *SBR_NO2*.

MBBR Anammox, case (MBBR_AMX)

The influent sludge liquor originated from the full-scale anaerobic digestion process at Bromma WWTP in Stockholm, Sweden. During the measurement period of 24 hours used for model calibration the ammonia load to the pilot-scale reactor was 70 g $N \cdot d^{-1}$ or 1.7 g $N \cdot (m^2 \cdot d)^{-1}$. This load corresponds to, compared to other periods, a low load and the amount of biomass in the system should therefore not have limited the total N removal efficiency (88%). The pH and temperature were relatively constant at 7.1 and 25 ºC, respectively.

The simulated amounts of biomass in the bulk and biofilm are shown in **Fig. 3A**. X_{AMX} dominates and is present throughout the entire biofilm. The process is intermittently aerated 45 out of 60 minutes (**Fig. 3D**) and X_{AMX} therefore has the possibility to grow also in the outer layers. X_{AOB} and a small amount of X_{NOB} are also present in the outer layers. Note that a not insignificant amount of biomass is found in the bulk water volume (shown as dots in **Fig. 3A**). In the model, and according to experimental observations, there is heterotrophic activity in the system as well.

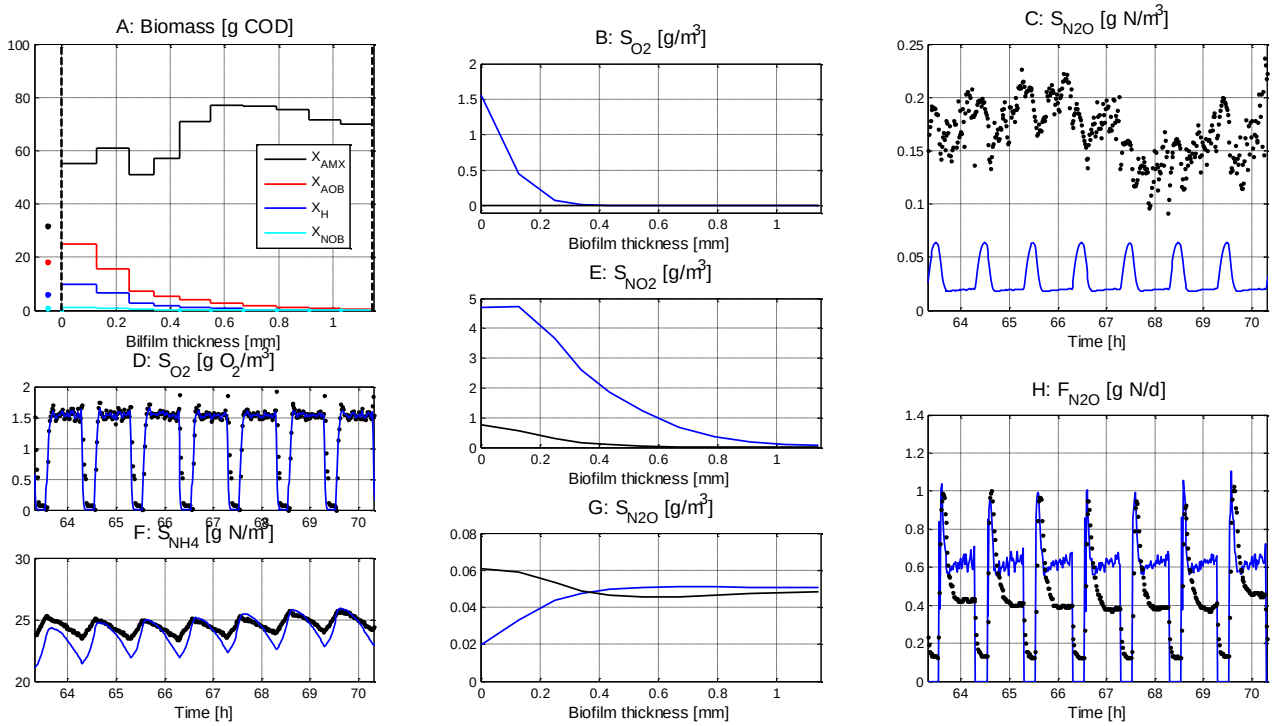

Figure 3. Scenario *MBBR_AMX*. **A:** Simulated amounts of active biomass in the biofilm (lines) and bulk (dots). **B, E, G:** Simulated concentration profiles during the end of aerobic (blue line) and anoxic (black line) conditions. **C, D, F, H:** Measured (markers) and simulated (solid lines) concentrations and mass flows.

Simulation results show that, compared to the previous case studies, the relatively low N_2O emissions of 0.5% during the studied period can be explained by heterotrophic denitrification. In the demonstrated simulations (**Fig. 3**) the ASMN default parameters for $X_{B,H}$ were used. Approximately 3% of the influent S_{NH4} is converted via nitrification and heterotrophic denitrification and 20% out of this amount is accumulated as S_{N2O}, probably because of low S_S concentrations from hydrolysis of particulates in the biofilm. The resulting emissions of N_2O are similar to the measurements and therefore, to simplify, AOB denitrification is not considered. It should be noticed that higher emission rates of N_2O were measured during other periods of the measurement campaign, which might indicate AOB denitrification. The reader is referred to Yang *et al.* (2013) for further information.

Fig. 3F shows the simulated and measured bulk S_{NH4} concentrations. S_{NO3} varied between 60 and 65 g $N \cdot m^{-3}$ and both S_{NH4} and S_{NO3} fully penetrate the modelled biofilm. **Fig. 3 B, E & G** show simulated concentration profiles from the bulk water through the biofilm layers at two occasions. The black line shows the profile after 15 minutes of anoxic conditions in the bulk while the blue line shows concentrations after 45 minutes of aeration. From these results it can be seen that S_{NO2} (**Fig. 3E**) is produced in the outer layer during aerobic conditions and penetrates almost the entire biofilm. When aeration is turned off, S_{N2O} is consumed by X_{AMX} and $X_{B,H}$. During anoxic conditions, S_{N2O} diffuses into the biofilm where it is converted by heterotrophic denitrifiers. As aeration is turned on S_{N2O} in the bulk volume decreases due to stripping and the diffusion changes direction so that S_{N2O} moves from the biofilm to the bulk.

The simulated N_2O emissions are shown in **Fig. 3H**. Emissions were measured also during non-aerated phases, which were not considered in the model. The simulated and measured dissolved N_2O concentrations are shown as time-series in **Fig. 3C.** The measurement data do not show a clear pattern but occasionally it can be seen that S_{N2O} increase during anoxic conditions.

The simulated S_{N2O} concentrations are generally lower than the measured ones. Based on the implemented model it is difficult to calibrate this effect because F_{N2O} (which is quite well predicted) is proportional to S_{N2O} and k_La_{N2O}. Thus, if the measurements are correct, either the stripping/flux model (including the diffusion coefficients) or the estimated k_La_{O2} is wrong.

Applied parameter values

In Table 1 below, a summary of the parameter values for X_H and X_{AOB} are shown. The table is not complete but includes the values that differ from the original publications (Hiatt and Grady, 2008; Mampaey *et al.*, 2013) and show the major differences between the three case studies.

Table 1. Calibrated model parameter values for the three case studies.

	K_{S1-4}	K_{S5}	$K_{EtOH,5}$	$K_{I5,HNO2}$
	[g COD·m^{-3}]			[g N·m^{-3}]
SBR_N/DN	20	100	1	0.001
SBR_NO2	20	40	-	-
MBBR_AMX	20	40	-	-

	b_{AOB}	μ_{AOB}	$f_{DNT,A}$	Y_{AOB}	$K_{O,AOB}$	$K_{NH3,AOB}$	$K_{NH3,AOB,DN}$	$K_{HNO2,AOB}$	$K_{NO,AOB}$
	[d^{-1}]		[-]	[g COD·(g N)$^{-1}$]	[g O_2·m^{-3}]	[g N·m^{-3}]			
SBR_N/DN	0.23	2.00	0.12	0.18	1.0	0.053	0.368	0.003	0.06
SBR_NO2	0.23	2.00	0.05	0.18	1.0	1.100	7.000	0.001	0.06
MBBR_AMX	0.08	1.41	-	0.18	1.0	0.053	-	-	-

Conclusions

The implemented biological process model, together with physical models for the SBR- and MBBR-process can partly describe the N_2O emission data from the three case studies.

The AOB denitrification model, which was adopted from Mampaey *et al*. (2013), could be fitted quite well to explain the behaviour of the nitrifying/denitrifying SBR. For the *nitritation only* SBR a high correlation to the ammonia concentration had to be assumed and may indicate that the implemented model is not able to fully describe the dynamics of the real system.

The four-step denitrification model, which was adopted from Hiatt and Grady (2008), could be used to model accumulation of dissolved N_2O during anoxic conditions in the nitrifying/denitrifying SBR. To model N_2O emission reduction caused by addition of ethanol, an additional COD state variable had to be added.

The stripping/flux equation in the implemented model may be overly simplified. It results in sharp N_2O gas emission peaks that are not observed experimentally. For simulation of full-scale N_2O emission data in general, the retention time of the gas including the measurement devices could probably improve the conclusions that can be drawn regarding N_2O formation pathways.

The N_2O emissions from the studied MBBR anammox process data where satisfactorily simulated by assuming heterotrophic denitrification only. The results from this case are preliminary and other measurements, which have not been considered in the present study, may indicate that AOB denitrification occurs as well.

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